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7. PERFORMING ORGANIZATION NAMES AND ADDRESSES
University of California - Los Angeles
Office of Contract and Grant Administration
11000 Kinross Avenue, Suite 211
Los Angeles, CA 90095 -1406

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# RPPR Final Report
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**INVESTIGATOR(S):**


**Name:** PhD Kang Wang
**Email:** wang@ee.ucla.edu
**Phone Number:** 3108251609
**Principal:** Y

Organization: **University of California - Los Angeles**
Address: Office of Contract and Grant Administration, Los Angeles, CA 900951406
Country: USA
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Submitted By: PhD Kang Wang Email: wang@ee.ucla.edu
Phone: (310) 825-1609

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**Major Goals:** Interplay between spin-polarized current and magnetization has led to many core phenomena and applications in spintronics. On the one hand, the spin-polarized current, generated by filtering through a ferromagnet (FM) or by spin Hall effect (SHE) in heavy metals and/or surface spin- momentum locking in topological insulators (TIs), can provide efficient means for manipulation of magnetization orientation through the spin angular momentum transfer. On the other hand, the magnetization can also significantly influence the electrical transport of spin-polarized current in these structures. The most well-known is the giant magnetoresistance (GMR) in the stacked FM layers with magnetization parallel or antiparallel to each other, which has played a major role in all modern developments of spintronics. Another nontrivial magnetoresistance (MR), the so-called spin-Hall magnetoresistance (SMR) in heavy metal/magnet bilayers, arises due to the back flow of spin-polarized current into the heavy metal when the SHE-induced spin accumulation at the interface is collinear with the orientation of the magnetic layer, which reduces the resistance of the heavy metal layer due to the inverse SHE. Recently, another intriguing unidirectional spin-dependent magnetoresistance (USMR) has been identified in the bilayers composed of high spin-orbit coupling (SOC) material and magnet, such as the heavy metal/FM and the Ga1-xMnxAs structures. The USMR depends on the relative orientation of the current-induced spin accumulation at the interface and the magnetization direction of the magnetic layer, parallel or antiparallel, in which the MR of the bilayer is different. The USMR could be understood from the current-in- plane GMR model or the spin-orbit torque (SOT) induced electron-magnon interactions. Both the SMR and USMR have potential applications in the sensing technology to detect the magnetization orientation in high-SOC material/magnet bilayers.
Compared with heavy metals and semiconductors with high SOC, TIs exhibit much stronger SOC and inverted band structure in the bulk. Most importantly, TIs possess the unique spin-momentum locked Dirac fermions on the surface, which are expected to be more efficient for generating spin polarization/accumulation at the interface, and hence are more efficient for producing the USMR when coupled with magnetic materials. To further explore the USMR in TI-based structures for potential technological application, it is crucial to investigate whether the surface states or the bulk carriers contribute most to the USMR effect, and also systematically study the correlation between the USMR strength and the magnetism in the structure.

**Accomplishments:** We demonstrated the nonlinear unidirectional spin-dependent magnetoresistance (USMR) in the modulation-doped magnetic topological insulators. This USMR arises due to the interplay between the magnetic dopants' magnetization and the current-induced surface spin polarization, when they are parallel or antiparallel to each other in the TI material. By changing the dopants' position in the structure, we reveal that the USMR is mainly originating from the interaction between the magnetization and the surface polarized spins, not the bulk carriers. Furthermore, from the magnetic field-, the angular rotation- and the temperature-dependence, we highlight the correlation between the USMR effect and the magnetism in the structures. The large USMR vs.

current ratio, which is several orders of magnitude larger than those reported in other material systems, suggests that this nonlinear effect may be employed as an effective method in detecting the magnetization orientation in spintronic devices with 2-terminal geometry.

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**PARTICIPANTS:**

**Participant Type:** Graduate Student (research assistant)
**Participant:** Qinglin He
**Person Months Worked:** 12.00 **Funding Support:**
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National Academy Member: N
Other Collaborators:

**Participant Type:** Graduate Student (research assistant)
**Participant:** Lei Pan
**Person Months Worked:** 12.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

**Participant Type:** Graduate Student (research assistant)
**Participant:** Yabin Fan
**Person Months Worked:** 12.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

**Participant Type:** Graduate Student (research assistant)
**Participant:** Qiming Shao
**Person Months Worked:** 12.00 **Funding Support:**
Project Contribution:
International Collaboration:
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Other Collaborators:

**Participant Type:** Graduate Student (research assistant)
**Participant:** Xiaoyu Che

**Person Months Worked:** 12.00

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# Scientific progress and accomplishments

## Foreword

Recently, immense interest lies in introducing and manipulating ferromagnetism in topological insulators (TIs) through either doping with magnetic elements or proximity coupling to a strong ferromagnetic system [1-4]. This interest is mainly driven by the novel quantum effects resulting from the non-trivial topology in TIs. The Dirac fermion surface states of TIs are topologically protected, being robust against lattice defect perturbations, non-magnetic imperfections, and surface reconstructions. Breaking time-reversal symmetry in these systems with magnetic dopants, such as Cr, results in opening of a surface exchange band gap, giving rise to a finite Berry curvature which leads to an intrinsic anomalous Hall effect (AHE). Inside this gap, non-zero Chern numbers of ±1 arise, giving rise to a chiral edge mode which encircles along the boundary of the TI [5]. In these edge modes, back scattering is forbidden due to the chirality. Such effect enables dissipationless transport using quantum AHE in TI system, leading to many potential ultra-low power spintronic applications. Also, these unique features make TIs a distinctive and exciting platform for exploring the interplay between charge and spin in the family of topological matter.

We has successfully demonstrated that by using the Néel order in antiferromagnets (AFMs) the magnetic order of a Cr-doped TI thin film through interfacial exchange coupling [3]. More recently, we demonstrated another new effect, that is, unidirectional spin-dependent magnetoresistance (USMR), observed in a TI/magnetic TI heterostructure. The USMR effect was usually identified in bilayers composed of a high spin-orbit coupling (SOC) material and a magnet, which depends on the relative orientation of the current-induced spin accumulation at the interface and the magnetization direction of the magnetic layer. In our TI/magnetic TI heterostructure, we recently observed USMR effect, which could be understood using the current-in-plane giant magnetoresistance model. Understanding the USMR in TIs assists potential applications in the sensing technology that can detect the magnetization orientation in high-SOC material/magnet bilayers.

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## Statement of the problem studied

The research program in current period aims at a deep understanding of novel physics in TI-based magnetic heterostructures from the perspectives of the SOT and the fundamental relativistic SOC. In this report, we focus on the USMR effect observed in a TI/magnetic TI bilayer.

The interplay between the spin polarized current and the magnetization through spin transfer torque develops considerable interest in recent years [6]. On one hand, the spin-polarized current can be generated by filtering through a ferromagnet or by spin Hall effect (SHE) in strong SOC metal/magnet systems, and/or surface spin-momentum locking character in TI systems. By such a regard, the magnetization orientation can be manipulated via the spin angular momentum transfer. On the other hand, the magnetization can also significantly influence the electrical transport of spin-polarized current. There is an emergent MR effect observed recently in strong SOC metal/magnet system, called spin-Hall magnetoresistance (SMR), which is resulted from the back flow of spin-polarized current into the metal when the SHE-induced spin accumulates at the interface. In Sections (4-5) of this report, we reported an observation of a unidirectional SMR in a TI/TI magnetic heterostructure. Strikingly, this SMR was found to depend on the relative orientation of the current-induced spin accumulation at the interface and the magnetization direction of the magnetic TI layer. These novel MR effects observed in TI systems described above have potential applications in the realm of sensing technology for detecting the magnetization orientation in strong-SOC material/magnet bilayers.

## Summary of the most important results

Compared with heavy metal case, TIs exhibit much stronger SOC and inverted band structure in the bulk. Most importantly, TIs possess the unique spin-momentum locked Dirac fermions on the surface, as depicted in Fig. 1a, which are expected to be more efficient for generating spin polarization/accumulation at the interface, and hence are more efficient for producing the unidirectional SMR when coupled with magnets. To explore the unidirectional SMR in TI-based structures, it is crucial to investigate whether the surface states or the bulk carriers contribute most to this effect, and also systematically study the correlation between the unidirectional SMR strength and the magnetism in the TI layer. These results are mainly summarized as follows.

(1) Demonstration of the unidirectional SMR effect in the TI/magnetic TI bilayer structures

By controlling the magnetic profile in a TI structure, we aim to identify the origin of the unidirectional SMR effect. First, we have grown a TI/magnetic TI bilayer structure, $(Bi_{0.5}Sb_{0.5})_2Te_3$(3 nm)/$Cr_{0.16}(Bi_{0.54}Sb_{0.38})_2Te_3$(9nm), by molecular beam epitaxy, followed by patterning into micron-scale Hall bar devices by photo-lithography, as shown in Fig. 1b. Due to the spin-momentum locking of the Dirac surface states, when passing a longitudinal charge current through the device, a surface spin polarization will be induced as a result of the Fermi surface contour shift in the momentum space, as illustrated in Figs. 1c-d. When scanning the transverse magnetic field to flip the magnetic TI layer magnetization from along $x$ direction (antiparallel to the surface spin polarization, see Fig. 1c) to $-x$ direction (parallel to the surface spin polarization, see Fig. 1d), the bilayer will experience a MR change from a low-resistance state to a high-resistance state. Equivalently, this unidirectional SMR resistance can also be revealed by reversing the current direction while keeping a constant in-plane magnetic field.

Furthermore, when applying an AC current, $I=\sqrt{2}I_{AC}\sin(\omega t)$ along *y*-axis, the AC current alternates its direction and consequently the unidirectional SMR resistance can be manifested on the 2nd harmonic longitudinal resistance, $R_L^{2\omega}=-\frac{\sqrt{2}}{2}I_{AC}\mathbf{d}R_L/\mathbf{d}I$ . This measurement result is summarized in Figs. 1e-f. In this figure, we plot the measured 2nd harmonic MR $R_L^{2\omega}$ vs. the magnetic field applied along *x*, *y*, and *z* axes in the Cr:TI(3nm)/TI(9nm) bilayer and the reversed-order structure, respectively. It's interesting to found that, by sweeping magnetic field along *x*-direction, when the relative orientations of the Cr dopants magnetization and the TI surface spin polarization are changed from parallel to antiparallel, $R_L^{2\omega}$ reversed sign from a negative to a positive value, as shown in Fig. 1e, consistent with the unidirectional SMR scenario. In the other reversed-order sample, i.e. a TI(9nm)/Cr:TI(3nm) bilayer (Fig. 1f), the unidirectional SMR reverses sign since the current-induced spin polarization direction on the bottom surface of TI is opposite to that of the top surface. When the magnetic field is scanned along y or z-axis in both structures, no 2nd harmonic MR was observed (see Figs. 1e-f), which suggests that when the magnetization in the magnetic TI layer is perpendicular to the surface spin polarization.

(2) Angular dependence of the measured unidirectional SMR effect

The above measurements indicate that the unidirectional SMR effect has a strong angular dependence on the Cr dopants' magnetization orientation with respect to the TI surface spin polarization. Following this assumption, we have carried out the rotation experiment by rotating the external magnetic field with a constant amplitude of 3 T in the *xz*-, *yz*-, and *xy*-planes, as illustrated in Fig. 2a, and meanwhile measured the 2nd harmonic longitudinal MR $R_L^{2\omega}$. The applied AC current is 0.6μA. The results are presented in Figs. 2b and c for the

Cr:TI(3nm)/TI(9nm) bilayer and the reversed-order structure, respectively. In both structures, in the *xy*-plane, $R_L^{2\omega}$ shows cosinusoidal relation with the magnetization's azimuthal angle $\varphi_M$ (since the film is isotropic in the *xy*-plane, $\varphi_M$ is equal to the field's azimuthal angle $\varphi_B$), demonstrating that $R_L^{2\omega}$ is proportional to the magnetization's projection on the *x*-axis, $M_x$. Similarly, in the *xz*-plane, we confirm the sinusoidal relation between $R_L^{2\omega}$ and the magnetization's polar angle $\theta_M$ (since the films have certain perpendicular anisotropy, $\theta_M$ can be obtained from the magnetic field's polar angle $\theta_B$ by balancing the different fields). For a clear comparison, we also plot the normalized $M_x$ as functions of the field's angle in different planes in Figs. 2b-c, denoted by solid curves, and they fit very well with the normalized $R_L^{2\omega}$ data (for convenience, we plotted $-M_x$ in Fig. 3c for the reversed-order bilayer structure since $R_L^{2\omega}$ in it changed sign).

In summary, we have demonstrated the large unidirectional SMR effect in magnetic TI bilayer structures, which is due to the interaction between the topological surface spin polarization and the Cr dopants magnetization. We also explored the angular dependence of this unidirectional SMR effect in different planes, confirming that this nonlinear effect is proportional to the magnetization projection along the transverse direction (i.e., along *x*-axis). In the next research period, we will analyze the current-density dependence and dopants position dependence, by which the surface and the bulk contributions can be distinguished.

(3) Exploring the origin of the USMR effect

(a) Electric-current dependence of the USMR resistance

After demonstrating the USMR is proportional to the magnetization projection on *x*-axis, $M_x$, it is natural to assume that this effect is also related/proportional to the surface spin polarization strength, which is governed by the current density applied. Furthermore, it is also crucial to identify whether this effect is a surface effect or bulk-related effect by controlling the distance between the surface polarized carriers and the Cr magnetic dopants. With these thoughts, we have grown different modulation-doped TI samples, with the 3nm Cr-doped TI layer at different vertical locations in the heterostructure, as depicted in Fig. 3d insets. To examine the current-density dependence, we performed the 2$^{nd}$ harmonic rotation experiments in all the modulation-doped structures in the *xz*-plane, with the probing AC current's amplitude ranging from 0.1 μA to 0.6μA (r.m.s. value). For example, the obtained results for the (3nm)Cr-TI/(9nm)TI bilayer and its reversed-order structure are shown in Figs. 3a and b, respectively. For a fair comparison, we have plotted the re-scaled $R_L^{2\omega}/R_L^{1\omega}$ in these two figures. Here $R_L^{1\omega}$ is the first-harmonic MR. The rotating field used has a fixed magnitude of 3T. We found that the $R_L^{2\omega}/R_L^{1\omega}$ data systematically increase when the applied current becomes larger. To summarize the current-density dependence, we pick up the data measured at $\theta_B$=90$^{\text{o}}$ (*i.e.*, when magnetization is along the transverse *x*-axis), and plot them versus the AC current density $J_{\text{ac}}$ (peak value) in Fig. 3c for different structures. We can clearly see that the USMR exhibits a linear dependence on the current density in these structures in the low-current regime, which is reasonable because within the linear response model the surface spin polarization is proportional to the applied AC current. Moreover, for the (3nm)Cr:TI/(9nm)TI bilayer and its reversed-order structure, where the dopants are located near the surface states, we observe very large (and with opposite sign) USMR $R_L^{2\omega}/R_L^{1\omega}$ data, see Fig. 3c. However, for the other trilayer structures [the (3nm)TI/(3nm)Cr:TI/(3nm)TI, (6nm)TI/(3nm)Cr-TI/(3nm)TI, and (3nm)TI/(3nm)Cr-TI/(6nm)TI trilayers], where the dopants are located inside the bulk (*i.e.*, away from the surface states), the USMR effect is almost negligible. This indicates that the USMR effect is a surface effect arising from the strong interaction between the surface spin-polarized electrons and the Cr dopants magnetization.

(b) Dopants position dependence of the USMR resistance

More importantly, we found that the USMR vs. current ratio, defined as (which could serve as a quantitative measure of the USMR strength), reaches the largest value of $4.2 \times 10^{-6}\ \mathrm{A}^{-1} \cdot \mathrm{cm}^2$ in the (9nm)TI/(3nm)Cr:TI bilayer and $2.3 \times 10^{-6}\ \mathrm{A}^{-1} \cdot \mathrm{cm}^2$ in the (3nm)Cr-TI/(9nm)TI bilayer. The value is nearly 6 orders of magnitude larger than the number reported in heavy metal/FM structures [7, 8] and two orders of magnitude larger than that in the $Ga_{1-x}Mn_xAs$-based system [9], suggesting that TIs are ideal materials for producing surface spin-polarization and enabling the USMR effect. To elucidate the USMR strength in different modulation-doped samples, we plot in Fig. 3(d) the $\left(R_L^{2\omega}/R_L^{1\omega}\right)/J_{ac}$ ratio as a function of the dopants' location in the whole structure along the vertical direction $z$ (the origin point is defined at the center of the sample). We can clearly see that when the dopants are on the bottom surface or on the top surface of the heterostructure, the ratios have very large (but opposite) values, consistent with the surface spins-induced USMR scenario; on the contrary, when the dopants are close to the center of the sample (*i.e.*, in trilayer samples), the ratios are very small, close to zero but with finite positive values. The minimal USMR is obtained in the (3nm)TI/(3nm)Cr:TI/(3nm)TI trilayer, where the dopants are almost at the symmetrical position in the whole structure. The fact that in all these trilayer samples, no matter whether the dopants are relatively closer to the bottom surface states or to the top surface states, or at the exactly symmetrical point, there is always a finite positive USMR, suggesting that there exists certain small current-induced spin polarization inside the bulk which is caused most likely by the electronic band bending along the vertical $z$-direction. These bulk spin polarization should not be induced by the bulk SHE because the SHE is expected to induce opposite spin polarizations with respect to the central origin point $z$=0, and consequently results in USMR effect with different signs. However, in all these trilayer structures, the devices always exhibit very small but positive USMR ratios; see Fig. 3d.

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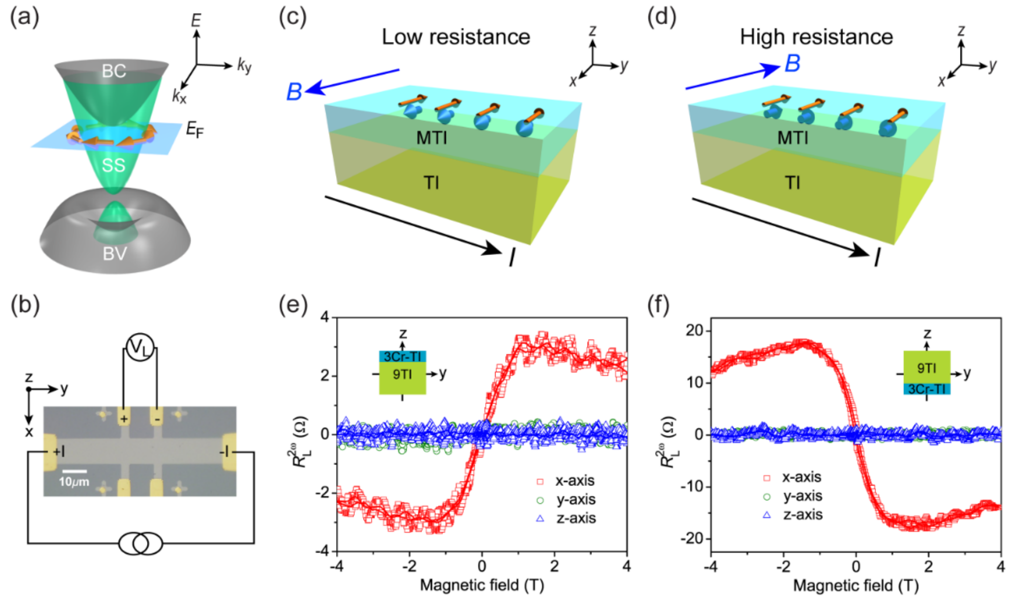


Figure 1

**Illustration of the unidirectional SMR effect in the TI/Cr:TI bilayer structures.** (a), Schematic of the topological Dirac cone on the top surface of Cr-doped TI, with the brown arrows denoting the spin-momentum locking direction. "BC", "BV" and "SS" stand for bulk conduction band, bulk valence band and surface states, respectively. $E_F$ is the Fermi level. (b), Microscopic image of the Hall bar device with illustrations of the magneto-resistance measurement set-up: current flowing from the left to the right (along *y*-direction) is defined as the positive current; $V_L$ measures the longitudinal voltage. The width of the Hall bar and the length between two neighboring Hall contacts are both 10 μm. (c) and (d), Schematic of the antiparallel and parallel orientation relations between the Cr dopants magnetization (blue arrows) and the surface polarized spins (brown arrows) in the Cr:TI/TI bilayer when passing a constant charge current *I* along the *y*-axis and meanwhile applying magnetic field along the *x* and -*x* directions, respectively. (e) and (f), Measured 2nd harmonic longitudinal magneto-resistance vs. magnetic field along different axes in the Cr:TI(3nm)/TI(9nm) bilayer and the reversed-order structure, respectively. The AC current applied was 0.6 μA (r.m.s. value). The measurements were performed at 1.9K.

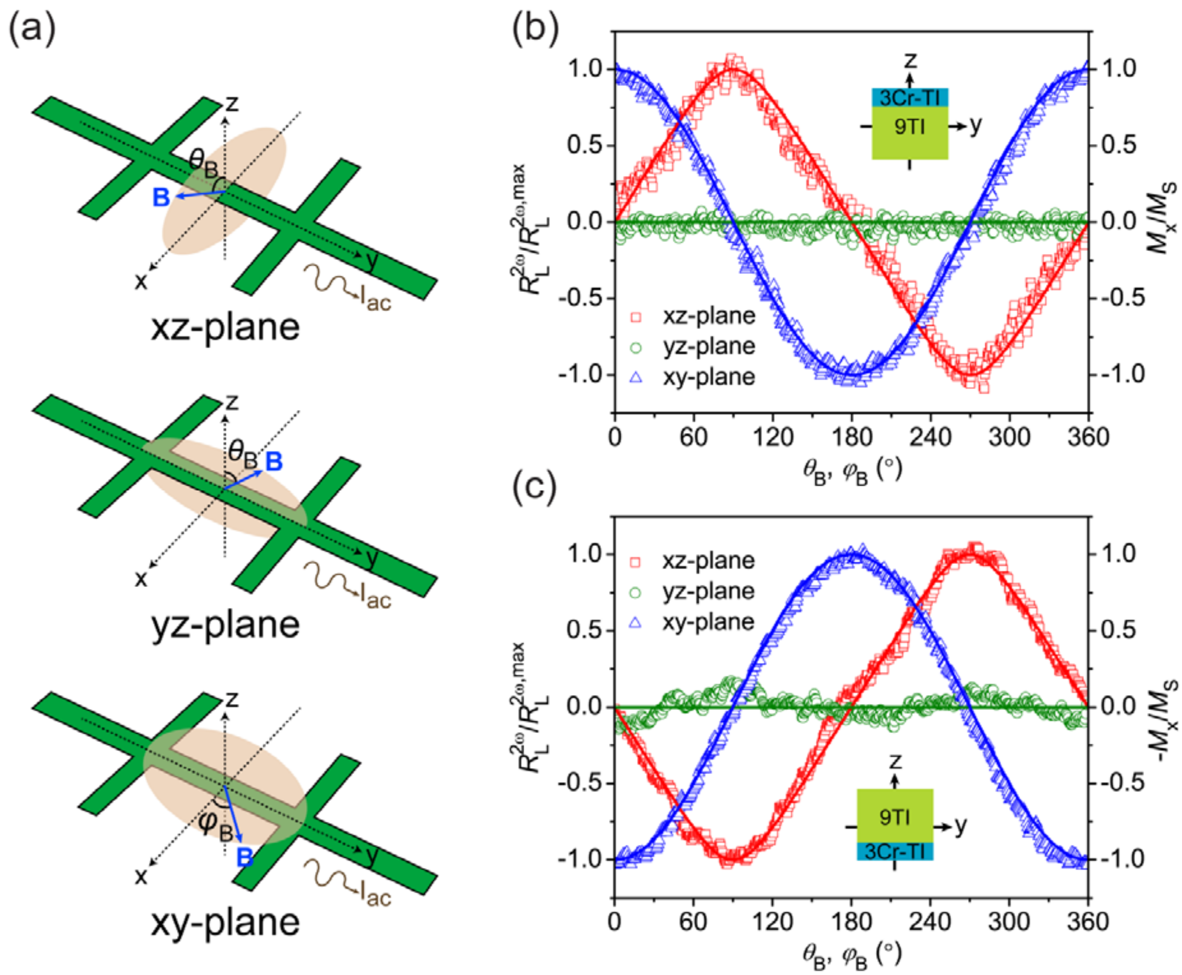


Figure 2

**Angular dependence of the measured unidirectional SMR effect.** (a), Schematic illustration of the rotational measurements in different planes. (b) and (c), Normalized 2nd harmonic longitudinal resistances as functions of the magnetic field rotation angle in different planes for the Cr:TI(3nm)/ TI(9nm) bilayer and the reversed-order structure, respectively. The magnetic field magnitude was kept at 3T and the AC current applied was 0.6 µA (r.m.s. value). The measurements were performed at 1.9K. The Solid curves show the normalized $M_x$ (in (b)) and normalized -$M_x$ (in (c)), and they fit well with the 2nd harmonic resistance data.

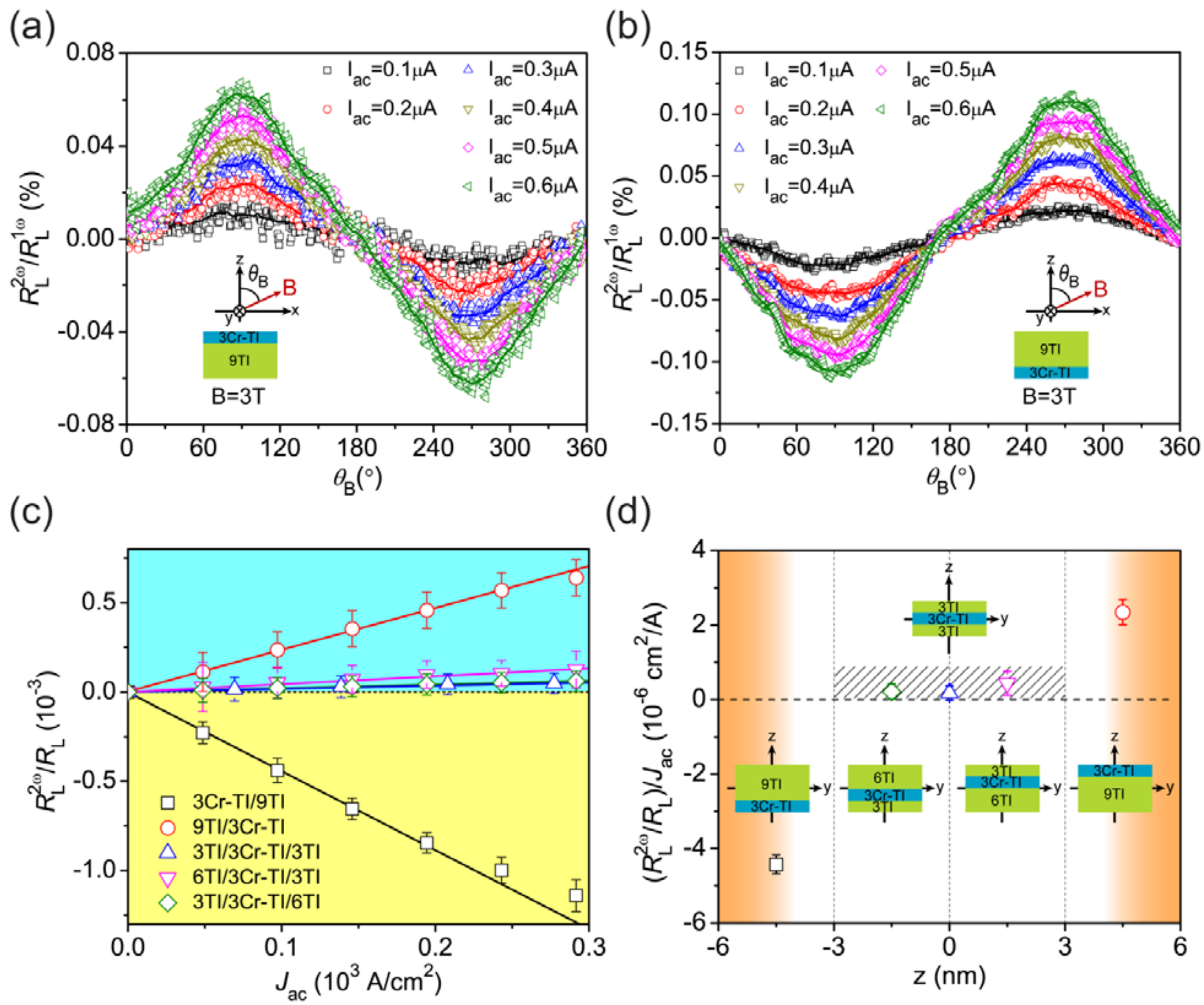


Figure 3

**Electric-current and dopants position dependence of the USMR resistance**. (a) and (b), 2nd harmonic longitudinal MR $R_L^{2\omega}$ as function of the magnetic field rotation angle in the *xz*-plane under different applied AC current $I_{ac}$ in the (3nm)Cr:TI/(9nm)TI bilayer and its reversed-order structure, respectively. The magnetic field magnitude is kept at 3T. (c), Re-scaled harmonic resistance, $R_L^{2\omega}/R_L^{1\omega}$, as a function of the AC current density (peak value) applied in different magnetic TI heterostructures. (d), The USMR ratio, defined as $\left(R_L^{2\omega}/R_L^{1\omega}\right)/J_{ac}$, plotted versus the Cr dopants position (center of the doped layer) in the modulation-doped TI structures. The shaded regions indicate the surface states on the two surfaces of TI, which are evanescent into the bulk.